

The impact of horizontal transport on the chemical composition in the tropopause region: lightning NO_x and streamers

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Abstract

The chemical composition of the region of the extra-tropical tropopause is influenced by chemistry and transport processes of either troposphere and stratosphere. On the basis of simulations with the coupled chemistry–climate model ECHAM4.L39(DLR)/CHEM two specific chemistry–transport interactions are investigated aiming to determine their importance for the ozone budget in the tropopause region. First, the transport and chemistry of nitrogen oxides from tropical lightning are analyzed. Second, the impact of low ozone air masses transported from the tropics into higher latitudes by wave breaking events (so-called streamers) is investigated. The first mechanism leads to an increase of ozone in the order of 5–10%. This additional ozone is not only formed in the tropical upper troposphere and then transported into the lower-most stratosphere (LMS), but it is also produced in the LMS, where the production is small but the residence time is long. The second mechanism is only important in the area 100–30 hPa, where ozone is not solely chemically controlled. In the lowest stratosphere at around 30°N and 30°S ozone decreases by up to 80%. In the tropopause region ozone decreases by 30% in summer and 50% in winter. The analysis clearly shows that the interactions of transport and chemistry in the tropopause region are highly complex and a simulation of that area requests a reasonable representation of the troposphere and the stratosphere.

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1. Introduction

Ozone as a climate gas is of special interest at tropopause altitudes, where its climate sensitivity is at a maximum (Wang and Sze, 1980). The main ozone production region is in the tropics at around 10 hPa. The Brewer–Dobson Circulation transports these air masses to higher latitudes and lower altitudes. However, at tropopause altitudes, ozone may also originate from other source regions. In the tropics, lightning produces NO_x yielding an ozone production in the upper troposphere. Subsequent troposphere to stratosphere exchange has the potential to contribute to the ozone budget at the extra tropical tropopause (see Fig. 1). The existence of such exchange processes and the impor-

tance for the chemical composition has been widely reported by measurement campaigns (e.g., Hoor et al., 2002). At slightly higher altitudes (pressure levels between 100 and 30 hPa), but below the main ozone production area, transport to higher latitudes is reduced due to the sub-tropical barrier. However, wave breaking leads to exchange of tropical air masses to higher latitudes, which can be detected in measurements of various trace gases, like N_2O and O_3 (Offermann et al., 1999), leading to an increase of N_2O at mid-latitudes and decrease of O_3 . But do those transport–chemistry interactions have a significant impact on the ozone budget at tropopause altitudes? To give an answer to the question, we applied the coupled chemistry–climate model ECHAM4.L39 (DLR)/CHEM (E39/C), analyzed trajectories and assessed the impact by specific sensitivity simulations, where transport of NO_x and ozone is inhibited or reduced for certain occasions (see details below).

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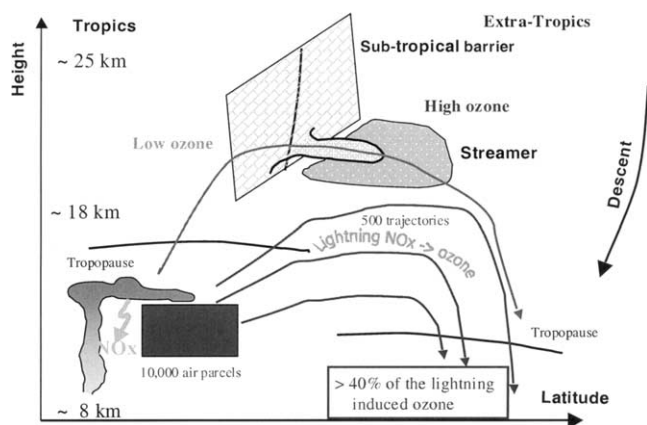


Fig. 1. Schematic diagram: zonal mean cross-section of the interaction of tropical lightning and streamers with mid-latitude ozone changes, as well as the method used to investigate these interactions. For details see text.

2. Chemistry–climate model E39/C

The coupled chemistry–climate model E39/C consists of the spectral atmospheric general circulation model ECHAM4.L39(DLR) (E39) and the chemistry module CHEM. E39 is based on the climate model ECHAM4 (Roeckner et al., 1996) with increased vertical resolution from 19 to 39 levels and the top layer centered at 10 hPa (Land et al., 1999). In this study, the model was applied with a horizontal resolution of T30 with a corresponding grid size of $3.75^\circ \times 3.75^\circ$. Water vapour, cloud water and chemical species are advected by a so-called semi-Lagrangian scheme. E39 includes parameterization schemes for small scale physical processes, like convection or cloud formation. The model has been used in a variety of tracer transport studies (Land, 1999; Land et al., 2002; Rogers et al., 2002; Timmreck et al., 1999).

The chemistry module CHEM (Steil et al., 1998) is based on the family concept. It contains the most relevant chemical processes for describing the tropospheric background NO_x – CH_4 – CO – HO_x – O_3 chemistry as well as the stratospheric homogeneous and heterogeneous ozone chemistry. CHEM includes 37 chemical species of which nine are explicitly transported and the others are grouped to three families. 107 photochemical reactions and four heterogeneous reactions on polar stratospheric clouds (PSCs) and on sulphate aerosols are considered in CHEM, however not yet bromine chemistry. Mixing ratios of methane (CH_4), nitrous oxide (N_2O) and carbon monoxide (CO) are prescribed at the surface. Nitrogen oxide emissions at the surface (natural and anthropogenic sources) and from aircraft are considered. Lightning NO_x emissions are calculated interactively depending on the mass fluxes in deep convective clouds according to Grewe et al. (2001).

Model climatologies and detailed descriptions are given in Hein et al. (2001) and Grewe et al. (2001). The

model has been applied for various chemistry–transport and climate–chemistry investigations (Schnadt et al., 2002; Grewe et al., 2001, 2002) and contributed to model inter-comparisons (e.g., Austin et al., 2003). The model also allows to calculate trajectories according to the modeled wind fields. Details are given in Reithmeier and Sausen (2002).

3. Lightning NO_x

To investigate the importance of tropical lightning NO_x a multi-step approach has been used (Grewe et al., 2002). First 10,000 particles were released in the lightning area (Fig. 2), which represents the main tropical upper troposphere region where tropical lightning emits nitrogen oxides. From that a main transport pathway has been detected. Second, simulations with and without lightning emissions were performed with the coupled chemistry–climate model. A second pair of simulations with and without lightning emissions were performed where the upper boundary for NO_y (=all nitrogen compounds) is lowered from 10 to 100 hPa to inhibit transport of lightning NO_y from the tropics to the mid-latitudes. Third, the simulated ozone production and destruction terms were interpolated to the mean trajectory to estimate the lightning ozone production areas by backward calculation.

From all trajectories which enter the northern lower-most stratosphere 70% show the pathway displayed in Fig. 2. To determine the pathway, all trajectories which pass the upper tropical troposphere and end in the lower-most stratosphere are selected. These trajectories are equally divided in a constant number of subsections. Based on those data points a mean trajectory is calculated, including a standard deviation (Fig. 2). Lightning changes ozone in the tropical upper troposphere by approximately 25% (Fig. 3, left) and around 10% at mid-latitudes. Since the nitrogen compounds emitted by lightning are transported to higher latitudes via tropo-

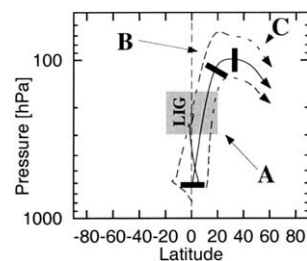


Fig. 2. Trajectory composites: 10,000 particles were released in the grey shaded area, 500 of them were transported into the lower-most stratosphere and 70% of these 500 followed the pathway. The dashed curves show the mean deviations of that pathway. A, B, and C mark areas with different chemical and dynamical characteristics (see text for details).

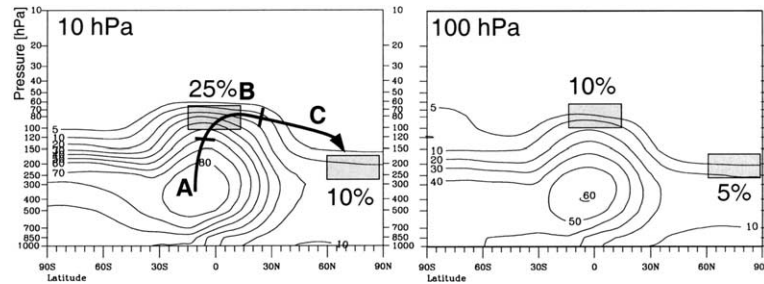


Fig. 3. Annual mean ozone changes (%) caused by lightning emissions simulated with the standard E39/C model (left) and with a lowered (from 10 to 100 hPa) upper boundary for NO_y (right). Differences between simulations with and without lightning emissions relative to the case without lightning are shown. Boxes are included at the same region to enable a better inter-comparison. The mean pathway from Fig. 2 is also included.

sphere to stratosphere exchange, this transport–chemistry interaction can partly be suppressed by lowering the NO_y boundary condition down to 100 hPa. The results (Fig. 3, right) clearly show a lower ozone change due to lightning. At the northern extra-tropical tropopause this reduces the lightning ozone increase from 10% to 5%. Troposphere–stratosphere interactions are of importance for at least 50% of the lightning induced ozone changes in the extra-tropical tropopause region.

By mapping the calculated ozone production and destruction rates to the mean trajectory three main areas with equal contributions were identified for the production of ozone due to tropical lightning (not shown). A (see Fig. 2) is an area with high ozone production rates but only low residence times, B is an area with medium ozone production rates and residence times, and C is an area where the lightning induced ozone production rates are fairly low, but the residence times are large. All three areas are of different chemical and dynamical characteristics, but contribute equally to the mid-latitude lightning induced ozone increase (Grewe et al., 2002).

4. Streamers

As discussed in the last section the transport of air masses from the tropics to higher latitudes has an impact on the chemical composition of the tropopause layer. Above the tropical tropopause large-scale transport into the extra-tropics is connected to wave-breaking events (streamers) due to the existence of the sub-tropical barrier. Below the main ozone production area, i.e., between 100 and 30 hPa, tropical air masses are characterized by less ozone than at mid-latitudes. Streamers therefore transport low ozone and lead to a decrease of ozone at mid-latitudes. Eyring et al. (2002) applied a meridional streamer detection criterion to simulated N_2O fields. N_2O as a long-lived species is a suitable tracer to detect transport characteristics. They compared 20 years of E39/C model data to nine years of ECMWF based KASIMA N_2O simulations and found a good representation of the seasonal cycle of the streamer frequency especially on the Northern hemisphere.

In a further simulation N_2O streamers are detected online. Whenever such a structure could also be detected in the ozone field, the ozone concentration is adapted to mid-latitude surrounding values. This approach leads to a suppression of the low ozone transport by streamers. It represents an additional ozone source at mid-latitudes. With this experimental design, it is possible to investigate the effect of ozone changes due to streamers on the chemical composition of the atmosphere. Fig. 4 shows results for DJF and JJA. The additional ozone source is centered at around 30° to 40° of both hemispheres at around 70 hPa (grey shaded area). The streamers show a pronounced seasonal cycle, which changes with altitude (not shown). Below 60 to 70 hPa more streamer mass is detected on the summer hemisphere, above that altitude streamers are mainly detected in the winter hemisphere, reflecting the Brewer–Dobson Circulation. The effect on ozone is quite large. The additional ozone source increases ozone by up to 400% above the tropopause in the tropics. Because of the decreasing chemical lifetime for ozone at higher altitudes, the impact is almost vanishing at 30 hPa

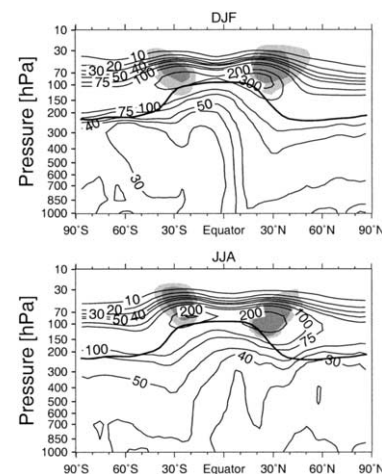


Fig. 4. Ozone changes (%) for DJF (top) and JJA (bottom) caused by an additional ozone source included in the model E39/C, which fills up ozone streamers according to the surrounding mid-latitude values. Differences are shown between a simulation with the additional ozone source and the standard simulation relative to the standard case.

although there is still a considerable additional ozone streamer source. At the extra-tropical tropopause ozone increases by approximately 100% in the winter hemisphere and 40% in the summer hemisphere for both hemispheres, respectively. And it still affects the tropospheric ozone content.

5. Summary and conclusions

In this paper interactions between the troposphere and the stratosphere, between transport and chemistry were investigated with respect to the ozone budget in the tropopause region. Two mechanisms were analyzed in more detail using simulations with the coupled chemistry–climate model E39/C. Clearly this study illuminates a part of those interactions, only. However, the importance of such interactions is clearly shown.

In the first part, we investigated nitrogen oxides emitted in the tropical upper troposphere by lightning. We found that the dominant pathway into the lowermost stratosphere is isentropic transport to higher latitudes around 100 hPa, i.e., above and below the tropical tropopause. On this way lightning induced ozone is equally produced (in terms of mass) directly in the source region, at around 100 hPa in the tropics and in the lowermost stratosphere. By including a lower upper boundary for NO_y in the model simulation, this transport was partly inhibited. The effect of lightning on ozone has been reduced by 50% at mid-latitude tropopause altitudes. It clearly shows that a simulation of lightning effects requires the inclusion of lower stratosphere dynamics and chemistry. Although there are clear indications that those transport processes are of importance (Hoor et al., 2002) there is still uncertainty about the amount of air masses transported to higher latitudes, which limits the quantitative findings, but not the qualitative statements.

In the second part, horizontal exchange from the tropics to higher altitudes (streamer) has been investigated at pressure levels between 100 and 30 hPa. They lead to a decrease of ozone at extra-tropical latitudes, which has been estimated to be 80% at around 100 hPa and 30° of both hemispheres and 30% (50%) at the extra-tropical tropopause of the summer (winter) hemisphere, respectively. It shows that for the simulation of ozone at tropopause altitudes and also for tropospheric ozone exchange processes between lower stratosphere tropical regions and mid-latitudes play a significant role. Since Eyring et al. (2002) showed that streamer events deduced from ECMWF data and from E39/C model simulation show a reasonable agreement, one can assume that the local impact on ozone, i.e., an ozone reduction, is reasonably simulated. The propagation of this perturbation depends on the quality of either transport and chemistry, which highly interact. This may lead to uncertainties of

the model results, but it is unlikely that the magnitude of the results is affected, since it has been shown that ozone chemistry (Hein et al., 2001) and the residual circulation is fairly well simulated by the model (Schnadt et al., 2002; Austin et al., 2003).

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